Detection of Concealed Special Nuclear Material Using Nuclear Resonance Fluorescence Technique

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***Abstract*–Detection method based on Nuclear Resonance Fluorescence (NRF) technique targeting concealed Special Nuclear Material (SNM) has been discussed. Customized DAQ system has been developed to handle high rate events in NRF measurements by implementing real-time DSP on fast ADC and FPGA chip. Experiments have been setup for feasibility study utilizing a 3 MeV Van de Graaff accelerator. NRF peaks from various samples have been observed using both HPGe detector and lanthanum halide scintillation detector.**

I. Introduction

THE ability to detect clandestine nuclear material has become a problem of contemporary importance and is being actively pursued by researchers globally. Because of the variability of possible environments, the detection and identification problem can be quite difficult. Passive and active approaches are being explored, using a wide variety of penetrating particles. One of the most interesting of these, and yet least investigated, involves Nuclear Resonance Fluorescence, the subject of this work.

NRF can be considered as the nuclear analog of the atomic X-ray fluorescence process [1]. NRF is the process of resonance excitation of certain nuclear levels by absorption of photons and subsequent decay of these levels by re-emission of equivalent radiation. Because the energy level structure is unique for each isotope, the resonant energies can be used as fingerprints for isotope identification. Thus, the energy spectrum of the resonantly scattered photons can be used to identify the detailed isotope content of materials of interest, allowing differentiation between U-238 and U-235, Pu-239 and Pu-240, for example. Furthermore, these nuclear emissions occur at high energies, typically 3-10 MeV, which makes them highly penetrating and difficult to shield.

One feature of NRF that makes it a powerful tool for isotope identification is the sharp profile of the NRF cross section [2]. The width of the cross section is typically of the order of 100 meV, while the peak value is 100's of barns for strongly resonant levels. The resonance will spread over an energy range of several eV owing to Doppler shifts caused by thermal motion of the nuclei. Thus, the peak value of the cross section will be reduced to a few barns. This is usually comparable to or larger than the cross section for photo-atomic reactions. Such a distinctive resonance makes it easy to detect even a small quantity of material of interest.

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Bremsstrahlung x-ray has become a widely used excitation photon source and is what we used in this work. The continuous energy distribution of bremsstrahlung sources overcomes all problems caused by recoil energy loss. However, the resonance spread (on order of eV) is about one millionth of the energy spread of a bremsstrahlung source. This means that less than one-millionth of the total photon flux contributes during the excitation process. Detectors used to measure the resonant photons have to have high energy resolution. The resonant photons are almost mono-energetic, with an energy spread (eV) thousands of times smaller than the best energy resolution that modern radiation detectors can provide (keV). These resonant photons sit on a continuous background, so the better the energy resolution of the detector, the better the signal-to-noise ratio. HPGe detectors provide the best energy resolution among commonly available radiation detectors. HPGe detectors with high detection efficiency are currently the best detectors for NRF experiments. In contrast, recently developed lanthanum halide scintillation detectors offer good energy resolution and a fast decay constant. The fast primary decay allows the application of these detectors at an event rate much higher than what HPGe detectors can handle. And the energy resolution is sufficient to detect strong resonances from certain isotopes. System performance with both HPGe detector and lanthanum halide scintillation detector ($LaCl_3$) is studied during this work.

II. Detection Method Using NRF Technique

In homeland security applications, detection systems should be able to not only detect the presence of SNM but also identify the specific materials. This can be done with the photofission technique based on differences in fission product yields [3], or possibly on differences in the time dependence of delayed neutrons and gamma-rays. However, these approaches either require complicated spectrum analysis, or dual or multiple beam energies, and the accuracy can easily be affected by attenuation due to surrounding materials.

Conventional atomic X-ray imaging systems provide gross sensitivity to atomic number. These systems are based on the principle that materials preferentially attenuate or scatter photons with specific energies determined by atomic composition. The binding energy of the most tightly-bound K shell electrons is on the order of 100 keV. The energies of the X-rays have to be small enough to investigate the atomic structure. However, X-rays with such low energies suffer high attenuation and cannot penetrate objects with high density. This limits the X-ray fluorescence technique to surface surveys. Increasing the energy of the interrogation X-rays will

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make them more penetrating, but the attenuation coefficients are simply a function of electron density at these energies, and the gross sensitivity to atomic number is lost.

In NRF technique, because the energy level structure is unique for each isotope, the resonant energies can be used for isotope identification. Since every nuclear isotope with an atomic number bigger than two has energy levels that can be excited by photon absorption, detection systems based on NRF interaction are not limited to SNM, finding wide application in material characterization, waste management, and other areas in addition to homeland security. Compared with atomic fluorescence, NRF photons have much higher energies. These multi-MeV photons are highly penetrating and easy to detect.

In a detection system based on NRF, interrogation photons are used to induce resonance excitations in nuclei, and the de-excitation photons are measured to generate an energy spectrum. The two most commonly used detection schemes are the scattering method and the self-absorption method. In the scattering method, materials of interest are placed in an X-ray beam, and detectors are located out of the beam using a backscattering geometry as shown in Figure 1. All but the resonantly scattered photons at this angle are expected to have low energies, owing to the nature of Compton scattering. Thus, resonant photons can be discriminated on the basis of energy. In the self-absorption method, the X-ray beam goes through the object, after which it strikes a sample containing isotopes of interest. A transmission detector is located downstream in the beam line to monitor the flux of the off-resonance photons. A radiation detector, called the "notch detector" is located out of the beam and is pointed at a sample of the isotope of interest in a backscattering geometry, measuring the resonant photons scattered by this sample. The system setup is shown in Figure 2 below. The count rate under the resonant peaks registered in the notch detector is proportional to the flux of the resonant photons hitting the sample. Whenever a preferential attenuation of resonant photons is observed, the conclusion can be drawn that isotopes of interest are present in the object. This is registered as a disparity between the flux of off-resonant photons and that of resonant photons registered in the transmission detector and the notch detector separately. In this paper, we are only showing experimental results from our scattering experiments.

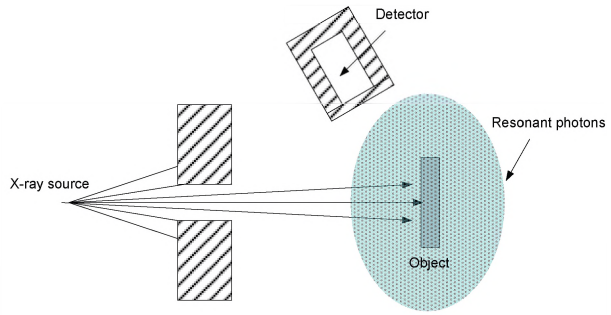

Fig. 1. Scattering detection method using NRF technique

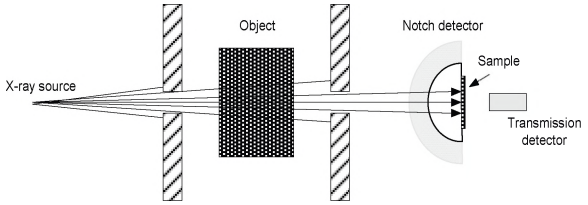

Fig. 2. Self-absorption detection method using NRF technique

III. Data Acquisition System

Not long ago, real-time digital signal processing could only be implemented on Digital Signal Processors (DSP). FPGA chips did not have the resources to implement complicated computation tasks like multiplication and division. In the middle 90's, a digital spectroscopic measurement system usually consisted of several DSP chips and a flash ADC, working at around 20 MHz [4]. However, FPGA's are well suited for this kind of application because of their distributed construction. Various tasks can be implemented on a single FPGA chip, running simultaneously without interfering with one another. Such a parallelism gives FPGA's the ability to handle much higher computational workloads than serial microprocessors. The results presented in this section show that nowadays a single, low-cost FPGA is powerful enough to drive an entire digital spectroscopic system.

This digital system was designed based on a fast ADC and an FPGA. The fast ADC continuously digitizes the input signal, and real-time signal processing as described below is implemented on the FPGA chip. Energy spectra are generated and stored in the memory available on the FPGA board and later transferred to a PC for display and analysis via the RS232 interface. Since signal processing is done in real time on the FPGA chip, the amount of data which need to be transferred between the DAQ system and the PC is much less than in the previous design, and a high-speed interface is not required.

A trapezoidal shaping method and a two-fold Moving Window Deconvolution (MWD) [5-6] algorithm were implemented on the FPGA chip for real-time DSP. The parameters can be adjusted to achieve a balance between speed and energy resolution.

The top-level system schematic is shown in Figure 3 below.

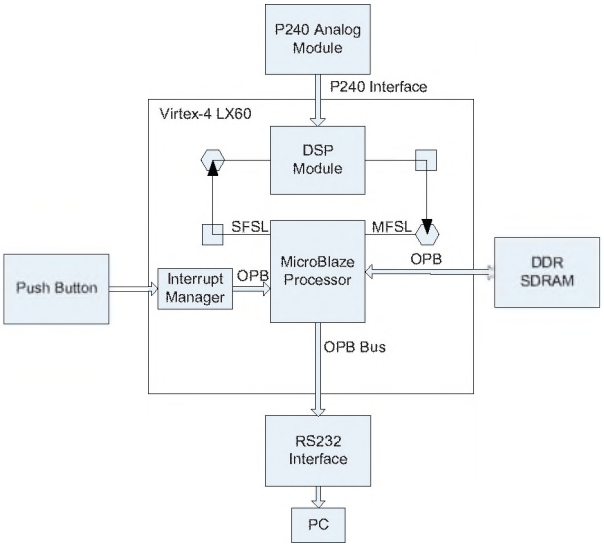

Fig. 3. Top-level System Schematic

Incoming analog signals were fed into one of the two analog input channels provided on the P240 Analog Module. After digitization, signals were passed to the Virtex-4 LX60 FPGA chip through the P240 interface. The ADC sampling rate was determined by the output clock frequency from the ICS 8442 clock synthesizer on the Virtex-4 LX60 development board. A DSP module was developed in Matlab and Simulink using the Xilinx System Generator. The design was then converted to an FPGA implementation. Digitized signals were processed within this DSP module. On the Virtex-4 LX60 FPGA, a MicroBlaze processor was implemented. This processor communicated with the DSP module through the FSL (Fast Simplex Link) bus. After digital shaping, energy information was extracted from the signals and stored in the DDR SDRAM. The MicroBlaze processor controlled the RS232 interface through the OPB (On-chip Peripheral Bus) bus. Energy spectra can be transferred to a PC through the RS232 interface, upon a user's request.

IV. Detectors and Experimental Setup

A series of NRF experiments were performed at the Radiation Laboratory of the University of Notre Dame. The lab is equipped with a Van de Graaff electron accelerator, with 100% duty cycle (DC beam) and an endpoint energy adjustable up to 3 MeV. The beam current in the DC mode can be set to as high as 200 μA. Bremsstrahlung x-rays generated by energetic electrons being stopped by a 1 mm thick tungsten converter were used as the interrogation source in these NRF experiments. Resonant peaks from various samples were sought using both a $LaCl_3$ detector and an HPGe detector.

In the experiments performed, two detectors were used to measure resonant photon spectra: a 1.5 inch by 1.5 inch $LaCl_3$ scintillation detector and an HPGe detector with 90% relative detection efficiency compared to a 3 inch by 3 inch NaI detector.

The $LaCl_3$ scintillation detector has a very fast decay constant of 28 ns. Together with the digital spectroscopic systems described in Chapter 3, it can measure energy spectra at a counting rate as high as 10^6 cps with no dead time and little degradation of energy resolution, as shown in section 3.2.3. However, because of its small size, this LaCl3 scintillation detector has fairly low detection efficiency. Its volume is only 1/8 of that of a 3 inch by 3 inch NaI detector. Thus, the solid angle it occupies is ¼ of what a 3 inch by 3 inch NaI would have at the same distance. An MCNP5 simulation showed that the intrinsic total efficiency falls to about 60 % of what it is for a 3 inch by 3 inch NaI detector for photons with energy around 2 MeV. The energy resolution of the $LaCl_3$ detector (4% at 662 keV) is high compared with other scintillation detectors, e.g. NaI detectors (6% at 662 keV). However, this resolution is a significantly worse than the ~0.2% HPGe detectors can provide. In NRF experiments, since the resonant photons have a very narrow energy spread while the background noise is continuous, signal to noise ratio (SNR) is inversely proportional to the energy resolution of the detector. Thus, the spectra measured by the $LaCl_3$ detector will have a SNR which is worse than that measured by HPGe detectors. In homeland security applications, the SNR is directly related to the measurement time required to achieve a certain false positive/negative error rate [7]. Thus, a degraded SNR means that a longer measurement time is required if a certain error rate is to be achieved.

On the other hand, HPGe detectors offer the best energy resolution among commonly available photon detectors. The one used in our NRF experiments is the ORTEC GEM90. It offers a relative detection efficiency of 90% when compared to a 3 inch by 3 inch NaI detector. The energy resolution is claimed to be 2 keV at 1.33 MeV. However, the best resolution that was obtained with our unit is 3.2 keV at 1.33 MeV.

The disadvantage of using HPGe detectors in NRF experiments is the slow processing time for each event. Since the shape of the output signal from the HPGe detectors varies according to the interaction position, the time constant of the shaping amplifiers used with HPGe detectors is usually set to a value close to the maximum of the charge collection time, which is of the order of a microsecond for a crystal as big as the GEM90. Plus, conventional spectroscopic systems usually have a processing time of several microseconds following each event. With the pile-up rejection feature enabled, this limits the maximum throughput rate to less than 10^5 cps. In addition, the energy resolution (FWHM) of HPGe detectors rises roughly linearly with the count rate up to around 10^5 Hz [8]. As discussed above, good energy resolution is essential in NRF experiments. In order to get an adequate SNR, a common rule is to keep the dead time below 10% in each measurement. This results in a maximum counting rate of 10^4 cps for our system (ORTEC GEM90 detector and ORTEC 926 MCA).

The schematic of the experimental setup is shown in Figure 4.

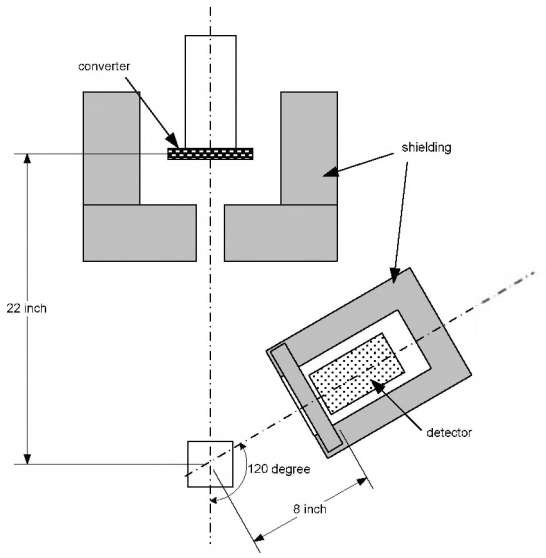

Fig. 4. Schematic for the scattering experiments

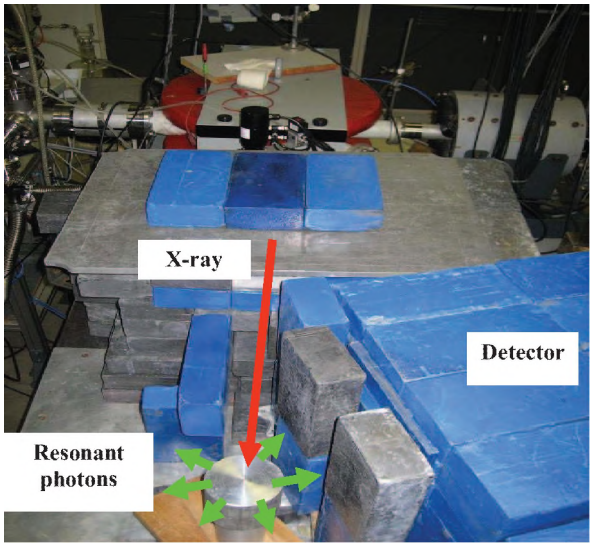

Fig. 5. The actual setup for the scattering experiment

In these experiments, the bremsstrahlung x-rays generated by the 3 MeV Van de Graaff accelerator were collimated down to a 2 by 2 inch beam. The scatterer is located in the beam, 22 inches away from the bremsstrahlung converter. The detector is shielded by 8 inches lead on each side, and 6 inches on top and bottom. A 1 inch thick lead sheet was put in front of the detector to block low-energy photons. Power supply and signal wires ran through the back of the detector shielding. The shield in the backward direction was 4 inches of lead. The detector was located 8 inches from the scatterer, at a backscattering angle of 120 degree. Pictures of the actual setup are shown in Figure 5.

V. Measurement Results

Two 40-minute BN measurements were performed with the $LaCl_3$ detector. One measurement was made with the BN sample in the beam and the other without. The results are shown below in Figure 6.

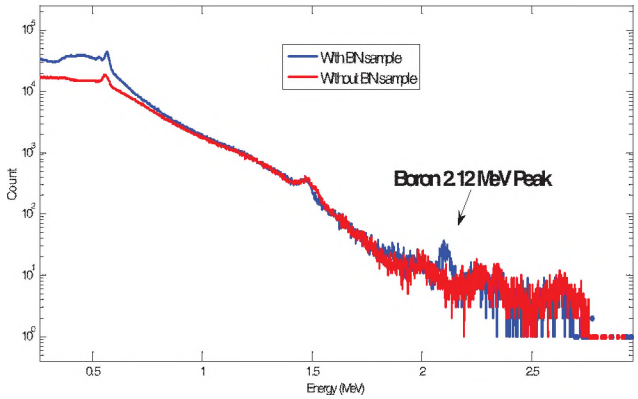

Fig. 6. Measurement results from NRF experiments using a BN sample

Two 40-minute Al measurements were performed with the $LaCl_3$ detector. One measurement was with the Al sample in the beam and the other without. The results are shown below in Figure 7.

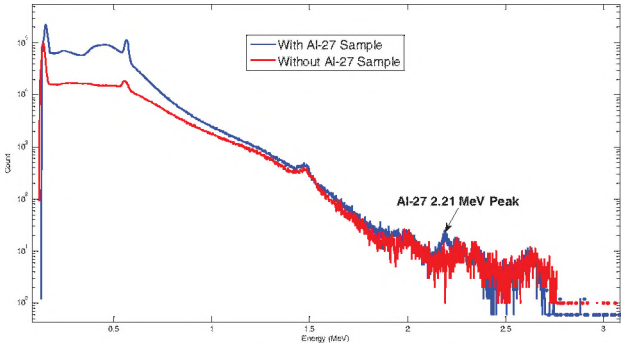

Fig. 7. Measurement results from NRF experiments using an Al sample

The following spectrum shown in Figure 8 for the BN sample was measured with a beam current of 20 μA, using the HPGe detector with the PUR function enabled. The measurement time was 10 minutes and the counting rate was only 854 cps. The energy resolution at 1.33 MeV was 3 keV. The net count rate under the photo-peak was measured to be 0.13 cps.

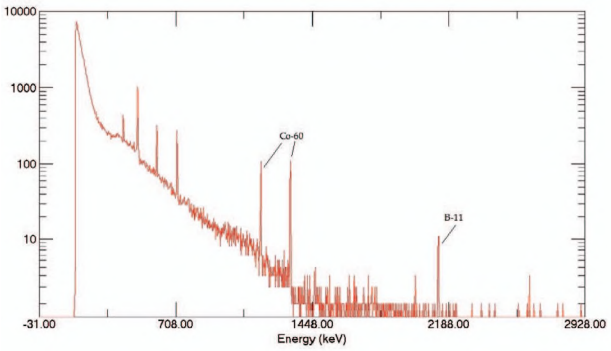

Fig. 8. Measurement results from NRF experiments using a BN sample

A 5-minute spectrum was measured with the Al sample using the HPGe detector. The result is shown below in Figure 9.

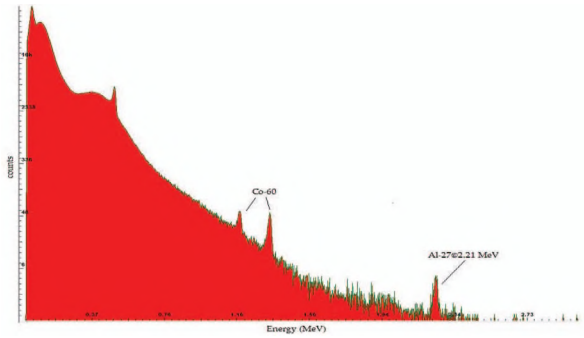

Fig. 9. Measurement results from NRF experiments using an Al sample

A measurement was performed with a thinner DU sample (3 inch by 3 inch by 0.1 inch slab) and PUR enabled. The count rate was 2.3×10^3 and the resolution at 1.33 MeV was measured to be 3 keV. The measurement time was 5.8 hours. Figure 10 shows the details of the region of interest (2.1 – 2.5 MeV).

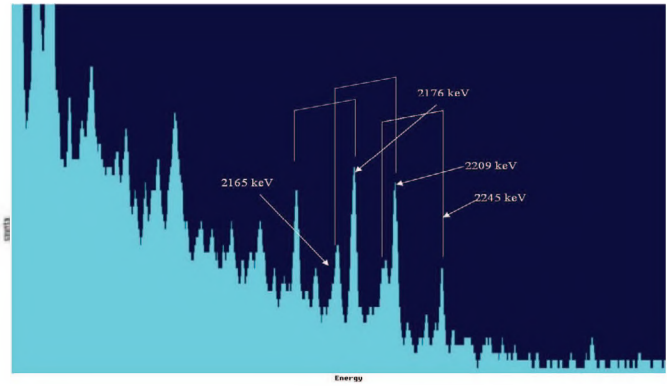

Fig. 10. Details of the ROI in the DU spectrum

VI. Conclusions

The detection method studied was based on nuclear resonance fluorescence technique. Resonance photons carry unique information of energy levels inside nuclei and can be used as signature signals for isotope identification. Scattering experiments were set up at the Radiation Laboratory of the University of Notre Dame. We used a 3 MeV Van de Graaff accelerator as our photon source. Using an HPGe detector, we were able to measure NRF peaks from various isotopes. For light materials, like B, Si and Al, the peaks are easily observable and the SNR was greatly improved when we implemented the PUR technique. NRF peaks from ^{238}U samples were successfully observed using the 90% efficient HPGe detector but required PUR, long acquisition times, and a thinned sample. The net count rate under the 2176 keV peak was measured to be 0.01 cps and the SNR was 49.6 ± 7%.

REFERENCES

[1] U. Kniessl, H.H. Pitz and A. Zilges, "Investigation of Nuclear Structure by Resonance Fluorescence Scattering," *Prog. Part. Nucl. Phys.* **37**, pp. 349, 1996

[2] H. Bethe, and G. Placzek, "Resonance effects in nuclear processes," *Phys. Rev.* 51, pp. 450, 1937

[3] D. Wehe, H. Yang, and M. Jones, "Observation of ^{238}U photofission products," *IEEE Trans. Nucl. Sci.*, Vol. **53**, no. 3, pp. 1430, 2006

[4] R. Belusevic, G. Nixon and D. Shaw, "An 80 Mbytes/s data transfer and processing system," *Nucl. Instr. and Meth. A* **295**, pp. 391, 1990

[5] A. Georgiev, and W. Gast, "Digital pulse processing in high resolution high throughput gamma-ray spectroscopy," *IEEE Trans. Nucl. Sci.*, Vol. **40**, no. 4, pp. 770, 1993

[6] M. Lauer, "Digital signal processing for segmented HPGe detectors preprocessing algorithms and pulse shape analysis," *Dissertation, Ruperto-Carola University of Heidelberg, Germany,* 2004

[7] J. Pruet, D. McNabb, C. Hagmann, F. Hartemann, and C. Bartly, "Detecting clandestine material with nuclear resonance fluorescence," *J. Appl. Phys.* **99**, pp. 123102

[8] J. Cooper, and D. Koltick, "Optimization of time and energy resolution at high count rates with a large volume coaxial high purity germanium detector," *Nuclear Science Symposium Conference Record, IEEE,* vol. **4**, pp. 2420, 2001